Aerosol/Cloud Condensation Nuclei (CCN) Closure during CRYSTAL-FACE

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Introduction

In 2002, as part of the multi-platform CRYSTAL-FACE field campaign, the CIRPAS Twin Otter carried a payload of aerosol and radiation instrumentation on twenty research flights in south Florida. The role of the Twin Otter was two-fold: to characterize the aerosol feeding the tropical convective cloud systems and to measure radiative effects beneath the cirrus anvils resulting from these systems.

The Twin Otter's payload included two continuous-flow cylindrical CCN counters, operating at supersaturations of 0.2% and 0.85%. The data collected during the campaign are sufficient for a detailed aerosol/CCN closure analysis, and indicate that in most cases, closure can be achieved even with an idealized chemical composition.

Also presented are data from flight CF-18, during which atypically high CCN concentrations were observed, with rapid fluctuations in concentration.

Laboratory Calibration

To verify the effective supersaturations of the CCN instruments, particles of known size and composition were simultaneously measured by it and a condensation nucleus counter.

The results of the verification experiments for one of the counters can be seen in the figure below. For both ammonium sulfate and sodium chloride, the data indicate a sharp transition between those particles too small to activate and those whose critical supersaturation is sufficient for growth in the instrument. Plotted as a function of critical supersaturation, the instrument's response for each species is found to be nearly identical, with the transition occurring at approximately 0.85%. Similar experiments with the other counter confirmed its effective supersaturation was 0.2%.
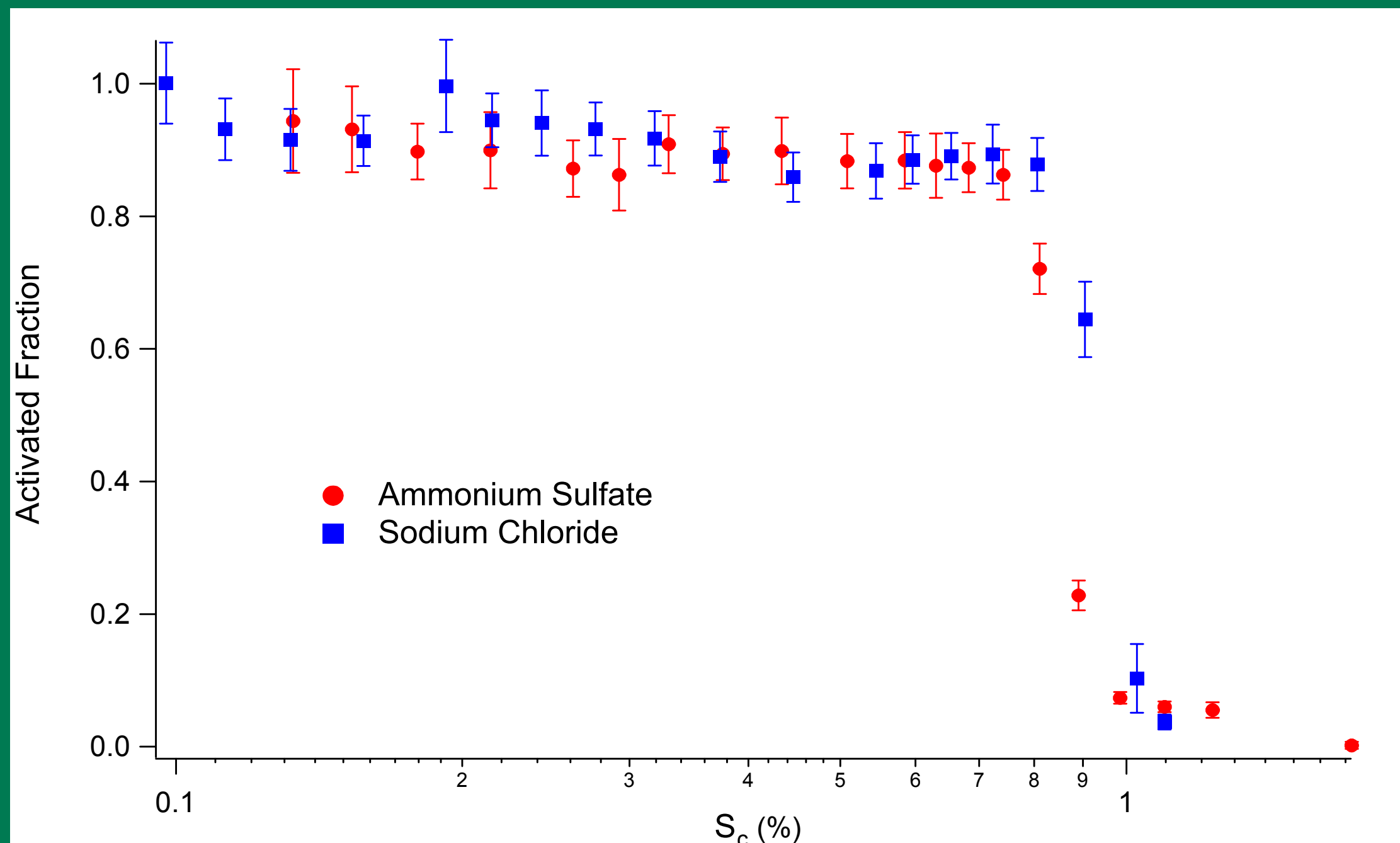
Instrument Intercomparison

To confirm that the CCN counters were operating as expected during flight, the temperature profile in one instrument was temporarily adjusted so that both counters were operating at S=0.85%. The results of this experiment are presented here. Over the course of 20-minute comparison, the mean ratio ($N_{Scripps}/N_{Caltech}$) was 0.917, with a standard deviation of 0.115. When the two brief periods of large disagreement are omitted, the mean increases to 0.929, with a standard deviation of 0.086.
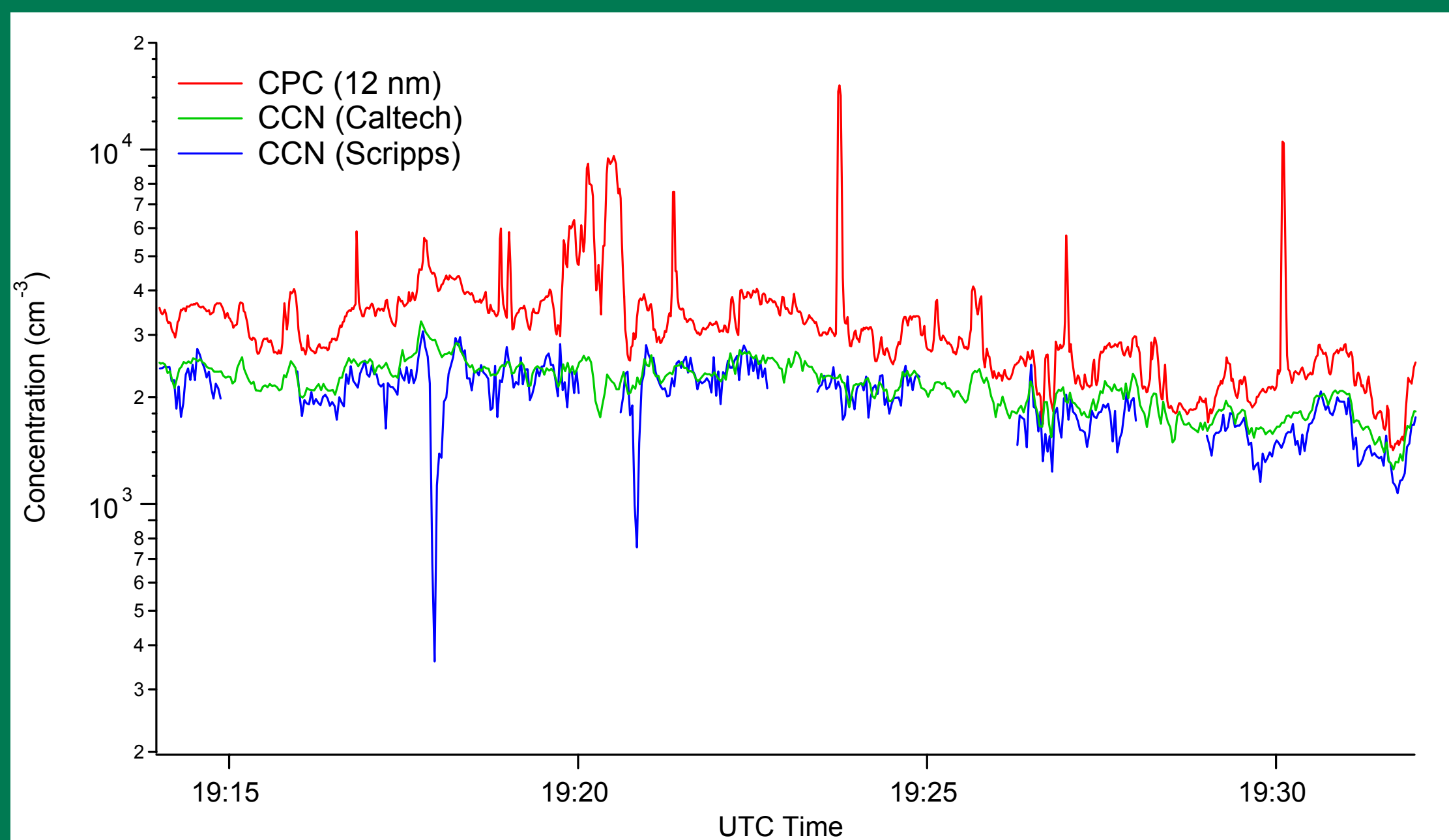
Field Observations

The tables and histograms below summarize the CCN observations made during CRYSTAL-FACE. At both supersaturations, the means are skewed upwards by a relatively small number of measurements at higher concentrations. The medians are 233 cm^{-3} at S=0.2% and 371 cm^{-3} at S=0.85%. Almost all of the high concentration measurements were made during the two flights on July 18 and the flight on July 28. The rest of the data are consistent with an air mass of marine origin.

S=0.2%

| Flight Number	Date	Number of Observations	Measured Concentration (cm^{-3})		Coefficient of Variation	
			Range	Mean	Range	Mean
CF-08	July 13	9	269-702	501	0.12-0.25	0.17
CF-09	July 16	36	129-582	391	0.07-0.27	0.13
CF-10	July 18	18	679-1553	850	0.10-0.49	0.22
CF-11	July 18	26	106-1310	649	0.10-0.76	0.36
CF-12	July 19	18	120-347	225	0.12-0.31	0.18
CF-13	July 19	25	39-80	55	0.25-0.76	0.46
CF-14	July 21	7	281-641	475	0.13-0.33	0.20
CF-15	July 23	55	33-304	141	0.12-0.68	0.28
CF-17	July 26	15	163-263	211	0.10-0.23	0.15
CF-18	July 28	52	219-1275	447	0.10-0.82	0.24
CF-19	July 29	28	50-261	109	0.13-0.43	0.25
CF-20	July 29	64	94-462	175	0.12-0.44	0.21
Overall		**353**	**33-1553**	**306**	**0.07-0.82**	**0.24**

S=0.85%

Flight Number	Date	Number of Observations	Measured Concentration (cm^{-3})		Coefficient of Variation	
			Range	Mean	Range	Mean
CF-1	July 3	45	324-1040	660	0.03-0.20	0.07
CF-2	July 3	57	288-801	514	0.03-0.99	0.09
CF-3	July 6	44	155-872	606	0.03-0.19	0.07
CF-4	July 7	29	399-935	554	0.03-0.41	0.22
CF-5	July 7	93	70-391	185	0.04-1.44	0.20
CF-6	July 10	14	427-851	614	0.04-0.23	0.08
CF-10	July 18	18	1138-2332	1413	0.03-0.19	0.07
CF-11	July 18	34	407-1661	1052	0.03-0.31	0.09
CF-12	July 19	20	287-640	456	0.04-0.20	0.08
CF-13	July 19	96	195-515	313	0.04-0.67	0.13
CF-14	July 21	52	225-1105	615	0.03-0.32	0.09
CF-15	July 23	84	218-720	326	0.04-1.28	0.13
CF-16	July 25	60	199-774	305	0.03-0.38	0.10
CF-17	July 26	27	261-402	314	0.03-0.43	0.09
CF-18	July 28	89	286-5999	1283	0.03-0.90	0.22
CF-19	July 29	38	84-436	215	0.04-0.50	0.16
CF-20	July 29	68	151-1193	385	0.05-0.56	0.15
Overall		**868**	**70-5999**	**533**	**0.03-1.44**	**0.13**
Omitting CF-18		**779**	**70-2332**	**447**	**0.03-1.44**	**0.12**

Aerosol/CCN Closure

Throughout CRYSTAL-FACE, measurements of the aerosol size distribution were made concurrently with the CCN measurements. For the closure analysis, CCN data were averaged to match the sampling interval (103s) of the differential mobility analyzer (DMA). An idealized ammonium sulfate concentration was assumed, and the predicted concentration was calculated by integrating the size distribution upward from the cut size measured in the laboratory calibrations. At S=0.2%, this cut size was 79 nm; at S=0.85%, it was 32 nm.

The results of the closure analysis are presented here. Each point represents one size distribution and the corresponding CCN measurements. A linear regression of the results, with the intercept forced to zero, result in a slope of 1.047 (R^2=0.911) when S=0.2%. When S=0.85%, the regression results in a slope of 1.201 (R^2=0.835); if flight CF-18 is omitted, the slope of the linear fit is 1.085 (R^2=0.770).

These overpredictions, ~5% at S=2% and ~9% at S=0.85%, are smaller than were found in all previous analyses using airborne measurements of CCN. The discrepancy is within the measurement uncertainty. More work will be required to determine whether the idealized compositional assumption can be used more generally.
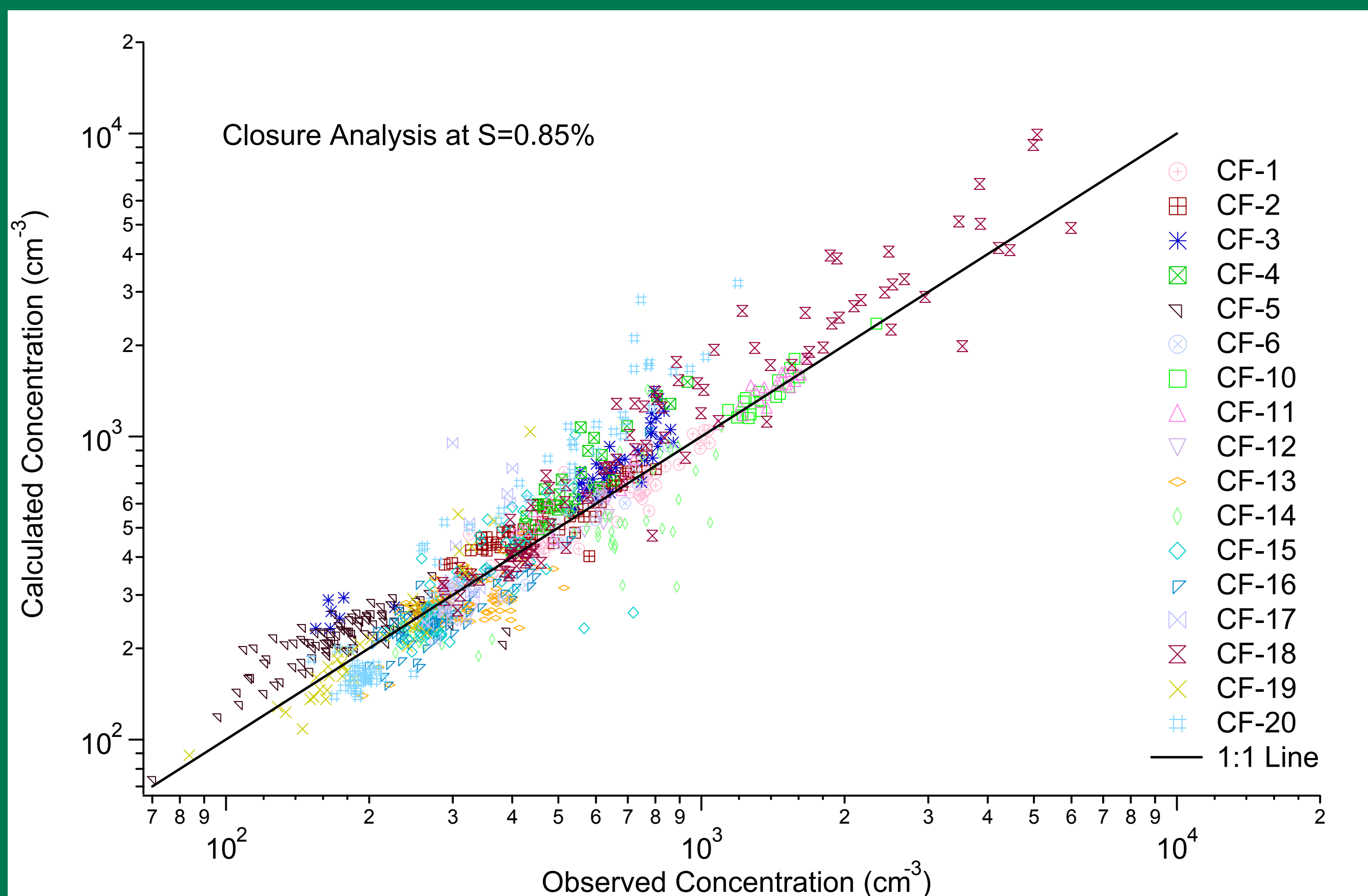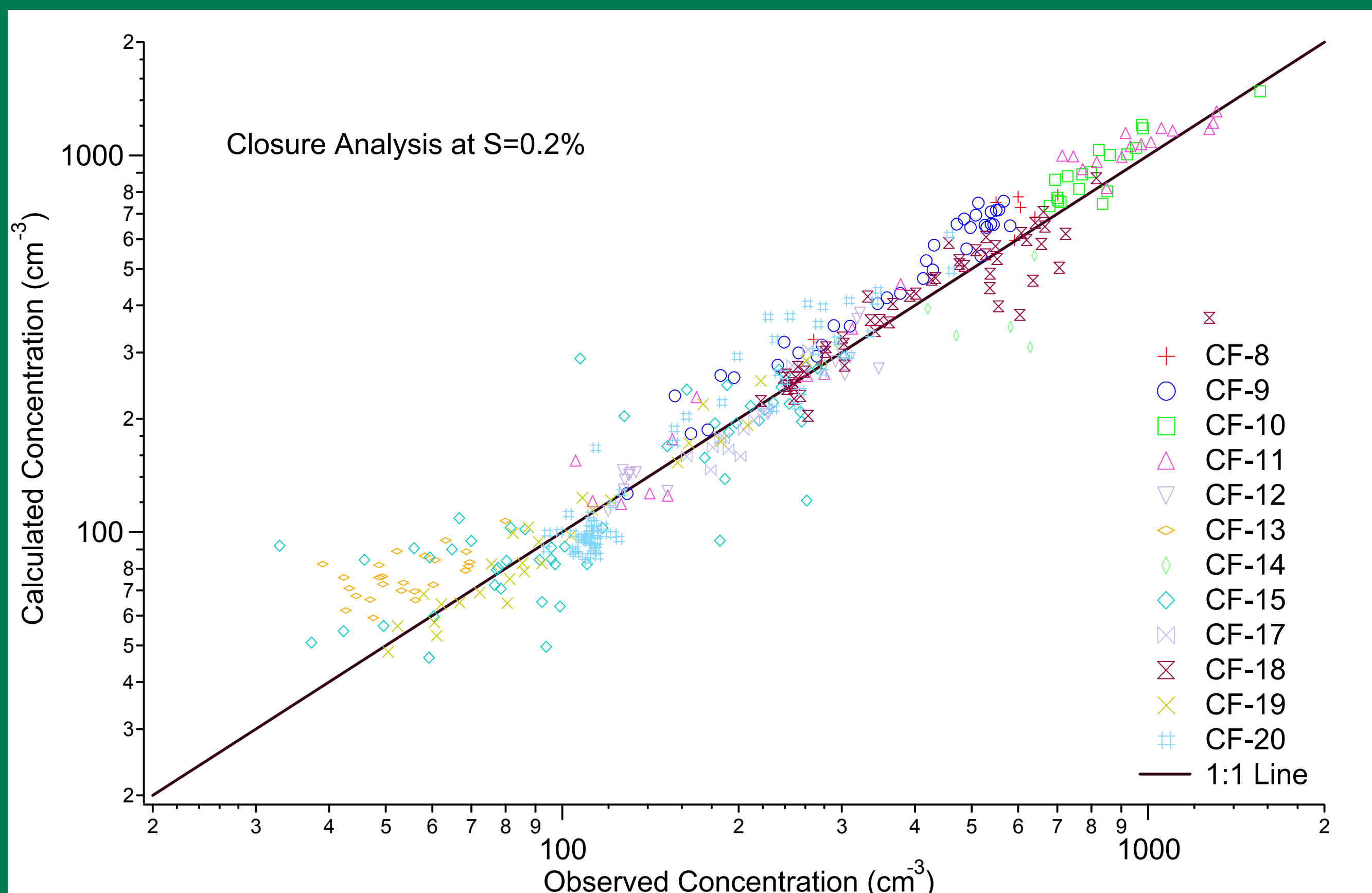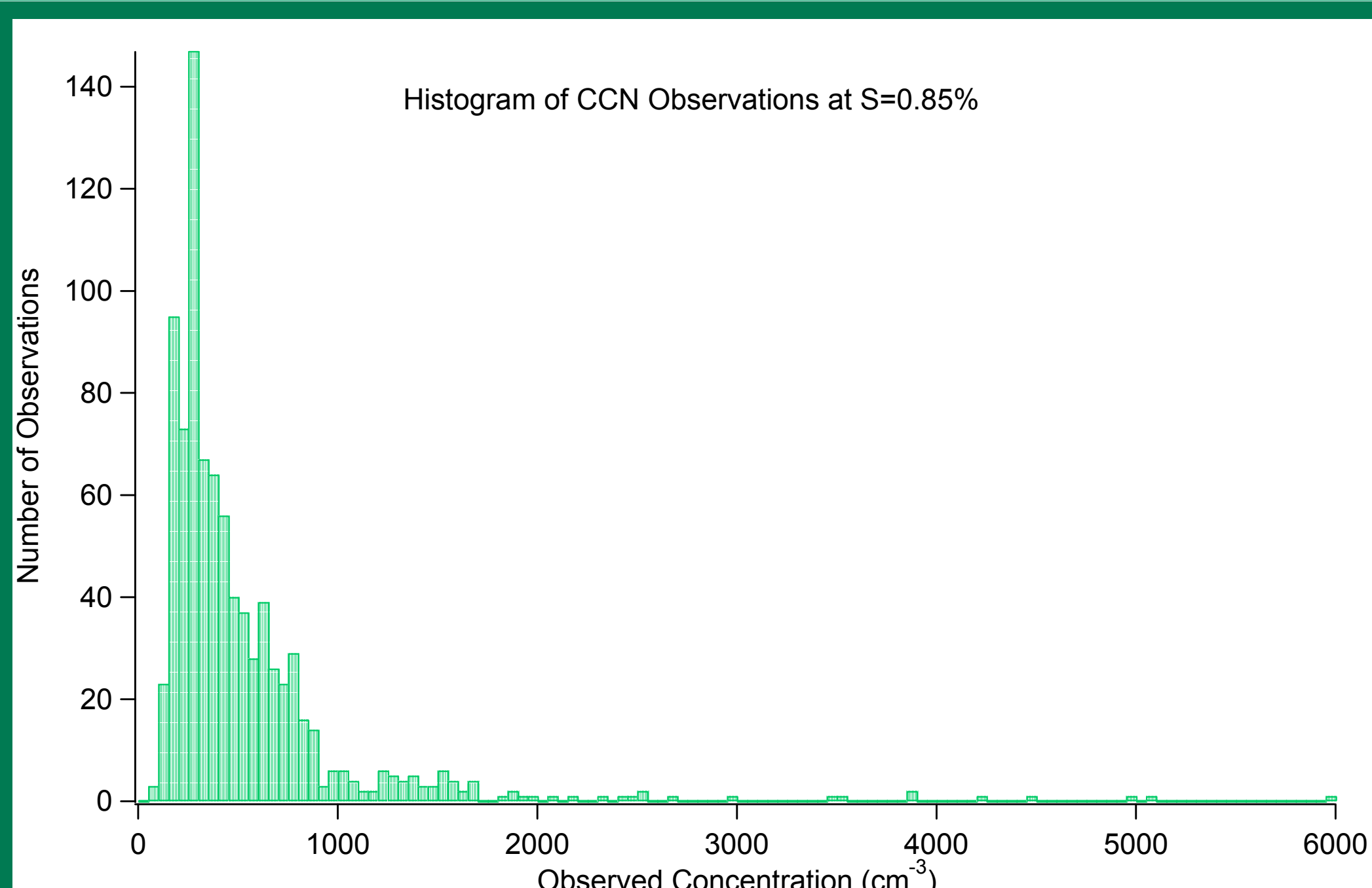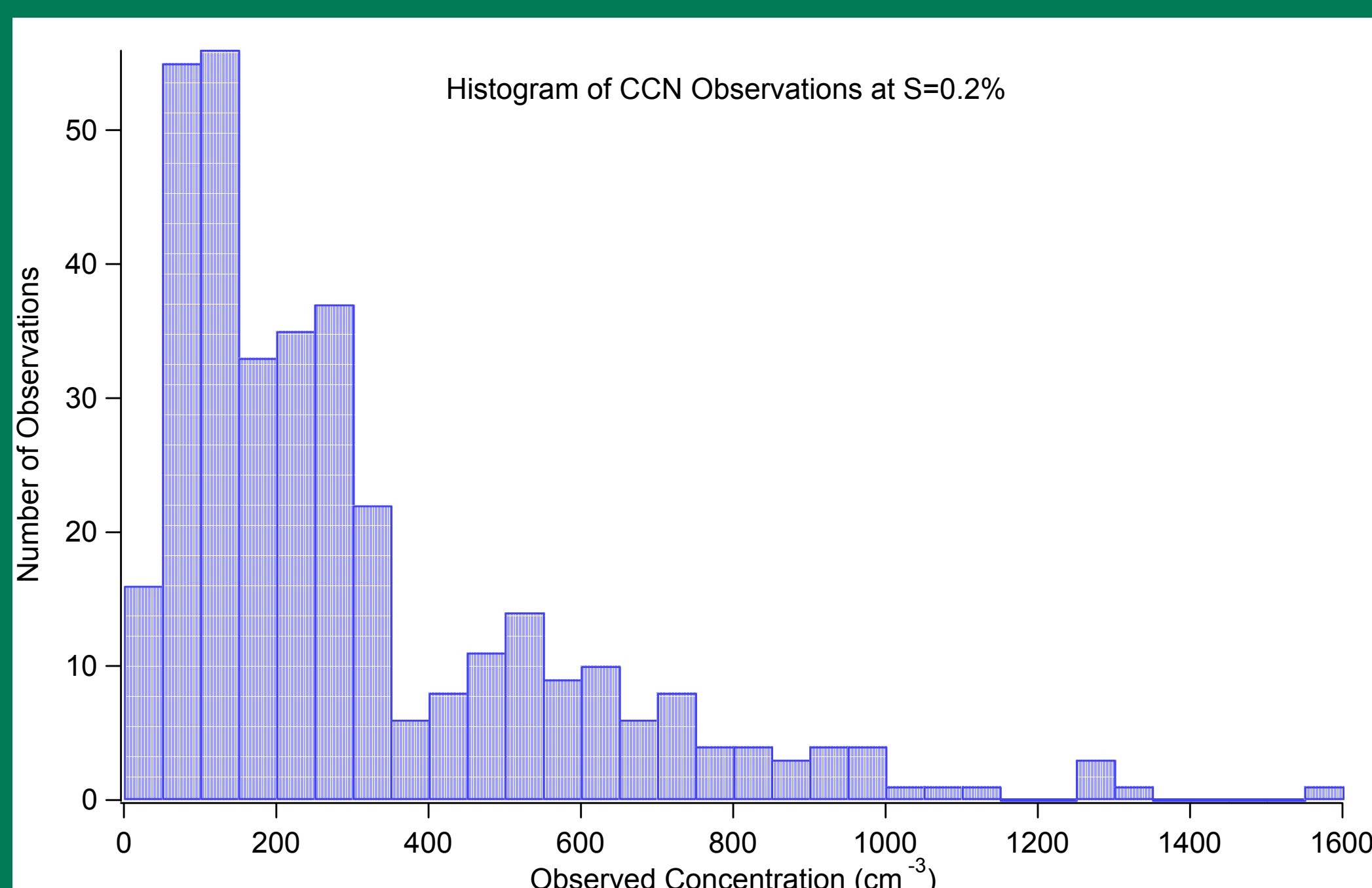
Case Study: July 28

On the July 28 research flight, CF-18, the Twin Otter encountered air masses with much higher particle concentrations than had previously been seen during CRYSTAL-FACE, as high as 30000 particles/cm^3. The sampled region was not well-mixed, as can be seen in the time-series data presented below; on several occasions concentrations changed by more than an order of magnitude in a few seconds.

The agreement between the predicted and measured CCN concentrations was at times very poor during this flight, particularly at S=0.85%. Size distribution data presented below indicate that the mode in the distribution often approached the predicted cut size for the CCN counter, and that nearly all the excess particles were below the cut size for the instrument operating at S=0.2%.
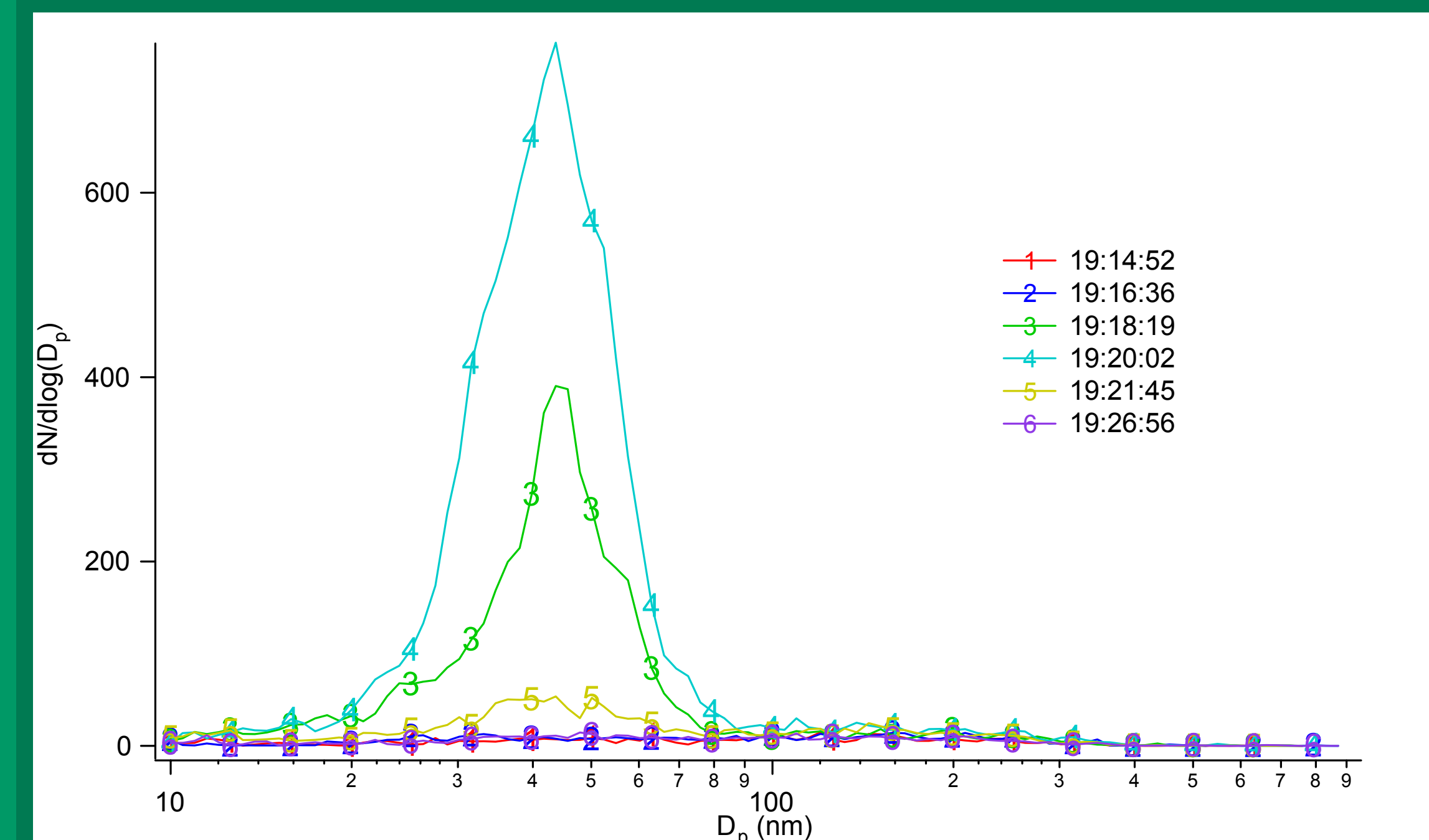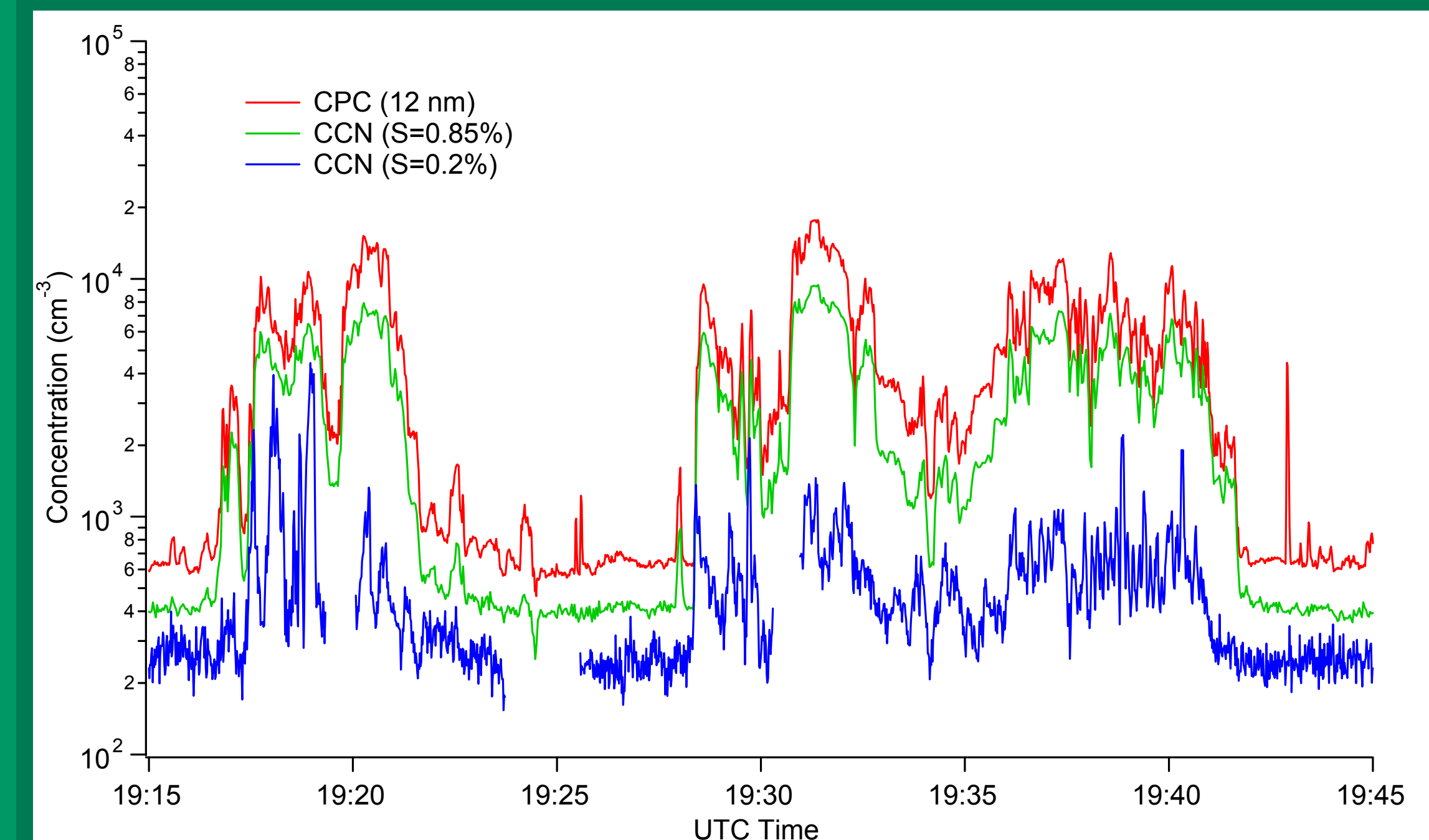
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Acknowledgments